Chapter 4

Assessment of Trends in Sediment Contamination Throughout the United States

Introduction

The historical reconstruction of sediment contamination serves as a tool to improve measures in reducing sediment contamination as well as to provide feedback to estimate the success of pollution control techniques and regulations. Sediments can provide an invaluable record of transformations due to natural or anthropogenic processes. These transformation processes have typically occurred over time periods greater than what has been monitored and assessed. Within a stable environment, the sediment retains a record of all contaminant inputs that can be dated on the basis of the decay of naturally occurring radioisotopes associated with the sediment (Hermanson, 1991). The sediment chronology can be established by approximate reconstruction of historical events. Some of the common methods used are the use of radioactive decay products such as ^{210}PB and ^{137}Cs, mineral magnetism, or correlation of sedimentary pollen or charcoal with historical records of logging or fires (Gubala et al., 1990). The depth distribution of the radioactive decay products in sediment cores provides valuable information on the period of sediment deposition. When the sediment cores are undisturbed, the activity of the radioactive product (e.g., ^{137}Cs) decreases exponentially toward the sediment-water interface (Bopp et al., 1998). By establishing a relationship between sediment depth and the time period, the history of the selected sediment components can be established.

One of the recommendations from the EPA report, *The Incidence and Severity of Sediment Contamination in Surface Waters of the United States*, Volume 1: *National Sediment Quality Survey* (USEPA, 1997) was to "consider whether to design future evaluations of NSI data to determine the temporal trends of contamination." In order to accurately assess potential trends in sediment contamination it would be necessary to measure levels of the same contaminants in sediment collected throughout time at the same randomly selected locations (not concentrating solely on "hot-spots"). This could enable identification of patterns or trends in sediment contaminants over time. Trends identified using this approach would however only be applicable to the areas where data were collected and could not be extrapolated beyond those areas. Extrapolation is not possible because of variability resulting from factors such as: land use patterns and historical anthropogenic activities. Despite the limitations imposed by the lack of routine monitoring information described above, EPA has developed an approach to provide the means for assessing changes in the extent and severity of sediment contamination over time for specific areas where sufficient data exist. To accomplish this, the data from the entire NSI database (data from 1980 through 1999) were evaluated. EPA's approach and the results of the Agency's analysis are discussed below. In addition to the trend assessment developed using the data from the NSI database, EPA has included information recently completed from the USGS National Water-Quality Assessment (NAQWA) program. In order to further evaluate potential trends in sediment contaminants, EPA has also conducted a literature review of studies conducted on sediment cores throughout the United States. This information is presented in Appendix E.

Database Trend Analysis

The NSI database contains more than 96,000 sediment samples collected from 41,594 sampling stations located in all 50 states, the District of Columbia, and Puerto Rico. These samples were collected from 1980 through 1999 and are associated with more than 3,000,000 chemical and other sediment-related observations. Data from just under one-half (19,470) of the stations were collected after January

1, 1990 and these stations were classified into tiers as described earlier. The purpose of this analysis is to determine whether the severity of sediment contamination in areas where data were evaluated is changing with time. For this purpose, all applicable sediment chemistry data from 1980 through 1999 were considered for analysis. The available data were gathered by a variety of heterogeneous monitoring programs. These monitoring programs provide an adequate amount of data for assessing sediment contamination at single sampling stations and times, however; these programs do not provide all the information needed for detecting trends in sediment contamination. Any of the factors described below can influence the analysis of trends.

Different/changing sampling and analytical methods over time. During the past 20 years, a wide variety of sampling and analytical methods have been used to collect and analyze sediment chemistry data. Different methods are often used by different monitoring programs. Programs take advantage of new technology as improved methods become available. Differences in monitoring program objectives (e.g., screening versus detailed assessment) also lead to different methods of sampling and analysis.

Limited quality assurance/quality control (QA/QC) information. Much of the historical data included in the NSI database is not associated with known data quality information. Although QA/QC and meta data are more frequently available for many datasets in the recent decade, it is still difficult, if not impractical, to apply more than a screening-level "use/don't use" approach for assessing quality of available datasets.

Monitoring strategies tend to focus on a single assessment. To detect temporal trends, it is usually appropriate to have collected a time series of data (multiple observations over time) at a station or to have two or more random samples, with each random sample representing a different time period. About two-thirds of the sampling stations (27,676) in the NSI database are represented by only one sediment sample, and only 740 stations are represented by 10 or more observations spread across at least a 5-year period. Only programs similar to the EMAP/REMAP design provide suitable random samples. At this time, however, the NSI database includes data from only 1,828 EMAP stations collected from 1990 through 1995, and the data are limited to the coastal regions of 19 states.

Despite these limitations, this report presents the methodology and findings of a temporal trend assessment of surficial sediment contamination throughout the United States for areas where data were evaluated.

Methodology

The following steps were used to prepare the data for analysis:

1. **Compute the predicted proportion toxic for all sediment chemistry samples collected from 1980 through 1999.** Computing the predicted proportion toxic using the logistic model (described in detail in Appendix B) is a convenient approach for combining measurements from several different chemicals into one value per sample. For those samples with only censored data (e.g., less thans and nondetects), the predicted proportion toxic was set to 0.10, 0.01 less than the minimum predicted proportion toxic from the logistic equation.

2. **Group samples based on latitude and longitude.** The latitude and longitude of each sample were rounded to the nearest 0.0001 degree. All samples from the same rounded latitude and longitude were then treated as if they had come from the same location. This allowed EPA to create data clusters, which have a time series of data at a single location.

3. **Average the predicted proportion toxic for a range of time periods** (1980-1983, 1984-1987, 1988-1991, 1992-1995, and 1996-1999). For each unique rounded latitude and longitude and time period, the average predicted proportion toxic was computed. (Each station could only have one value of predicted proportion toxic per time period.)

4. **Eliminate any data clusters where the predicted proportion toxic was not estimated in at least two of the time periods.** Based on a preliminary analysis of data contained in the NSI database, it was found that there were some major geographical shifts in data collection over the past 20 years. For example, during the early 1980s, a substantial amount of data was collected by the USGS in the Appalachian Mountains. In later years, data were not collected by the USGS in that region. This step eliminates the geographic diversity of the available data, but it also reduces the potential to compare geographically different data over time.

Application of this approach resulted in the availability of 4,153 stations for trend analysis. The locations of these stations in the contiguous United States are presented in the Figure 4-1. There are clear geographical biases in the available data. These biases range from little to no data in some states to wide-scale monitoring across an entire state. Because of the data preparation step, each station could only have one value of predicted proportion toxic per time period. Table 4-1 presents the number of predicted proportion toxic observations (or stations) by time period and hydrologic region. For example, there are 89 stations or predicted proportion toxic observations in Great Lakes hydrologic region (04) during the 1984-1987 time period. The location of hydrologic regions in the contiguous United States is presented in Figure 4-2. Hydrologic regions 19 and 20 represent Alaska and Hawaii, respectively. Because of the lack of data in hydrologic regions 01, 09, 10, 14, 15, 16, 19, and 20, individual statistics are not presented, but are included in national data where applicable.
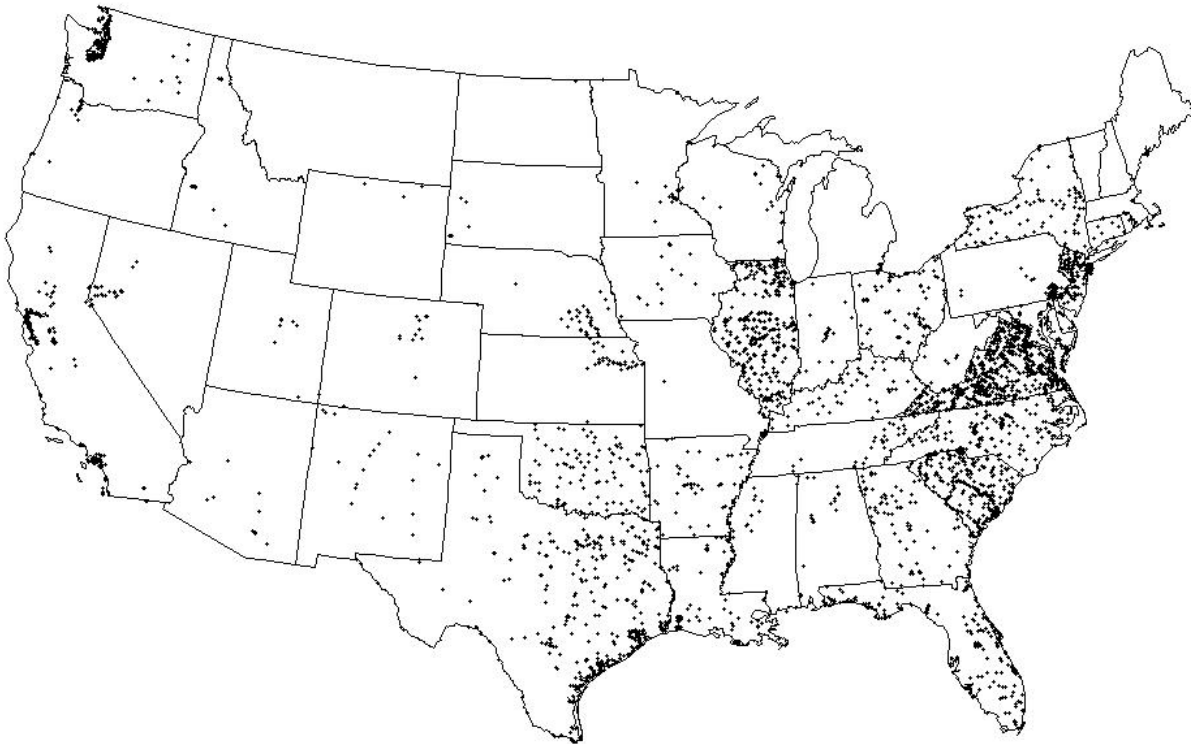

Figure 4-1. Locations of Data Clusters Used for Temporal Trend Analysis.

Table 4-1. Number of Predicted Proportion Toxic Observations Available for Trend Analysis After Data Preparation Step.

USGS Hydrologic Region	1980-1983	1984-1987	1988-1991	1992-1995	1996-1999
01	7	16	14	11	5
02	379	393	422	493	394
03	530	657	558	512	427
04	87	89	98	74	42
05	109	135	151	176	124
06	55	47	60	66	34
07	170	124	197	174	226
08	87	92	59	23	31
09	2	0	0	2	0
10	24	74	76	16	9
11	96	143	127	69	24
12	275	294	255	124	84
13	23	27	27	34	21
14	11	10	5	3	3
15	5	6	5	6	2
16	23	23	1	7	6
17	83	156	217	185	53
18	75	129	120	111	75
19	2	23	25	2	2
20	31	30	0	0	1

Hydrologic Regions

01: New England
02: Mid-Atlantic
03: South Atlantic-Gulf
04: Great Lakes
05: Ohio River
06: Tennessee River
07: Upper Mississippi River
08: Lower Mississippi River
09: Souris-Red-Rainy Rivers
10: Missouri River
11: Arkansas-White-Red Rivers
12: Texas-Gulf
13: Rio Grande
14: Upper Colorado River
15: Lower Colorado River
16: Great Basin
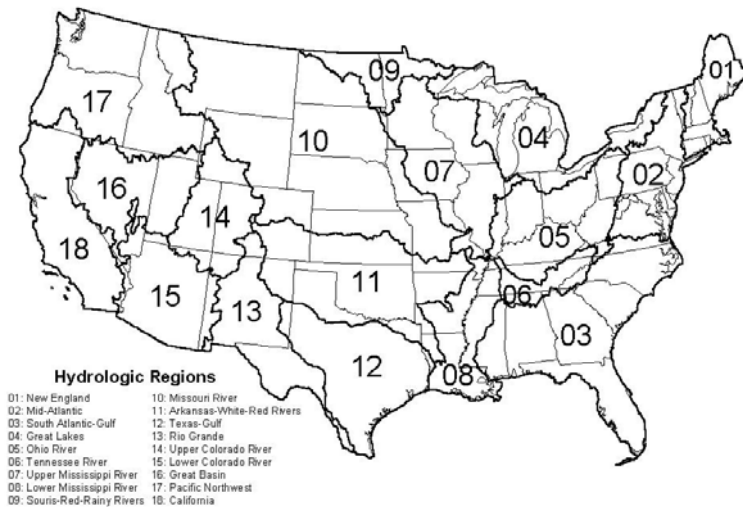17: Pacific Northwest
18: California

Figure 4-2. USGS Hydrologic Regions in Contiguous United States.

To evaluate the effect of focusing the attention on those stations with more concentrated data sets, the same data preparation step was performed using only the 740 stations associated with 10 or more observations spread across at least a 5-year period. Of these 740 stations, 422 stations have data for four or five time periods and 231 stations have data for three time periods. The station locations of these 740 stations are presented in Figure 4-3. Table 4-2 presents the number of predicted proportion toxic observations (or stations) by time period and hydrologic region. For example, there are 47

DRAFT - Do not cite, quote, or distribute December, 2001

stations or predicted proportion toxic observations in Mid-Atlantic hydrologic region (02) during the 1984-1987 time period.

Figure 4-3. Locations of Concentrated Data Clusters Used for Temporal Trend Analysis.

Table 4-2. Number of Predicted Proportion Toxic Observations Available for Trend Analysis After Data Preparation Step from Concentrated Data Clusters.

USGS Hydrologic Region	1980-1983	1984-1987	1988-1991	1992-1995	1996-1999
01	6	6	6	6	3
02	51	47	53	57	49
03	213	241	266	241	232
04	0	0	1	1	1
05	30	27	35	31	30
06	25	25	25	20	16
07	10	15	17	9	5
08	1	1	8	7	0
10	1	7	7	0	0
11	40	45	45	31	8
12	115	137	141	65	45
13	18	20	19	24	17
14	1	2	2	2	2
18	39	37	44	32	31
20	17	17	0	0	0

Results

The logistic model classification scheme described earlier in this document was used to classify stations (i.e., predicted proportion toxic greater than 0.5 is Tier 1). Table 4-3 presents the classification of observations by tier, time period, and hydrologic region for the data clusters. Overall, the percentage of Tier 1 or Tier 2 stations ranges from 54 percent during the first time period, to 47 to 49 percent during the middle three time periods, to 40 percent for the last time period. It is also possible to discern the results in Table 4-3 by examining the box plots in Figure 4-4. Figure 4-5 is a comparable box plot figure for the concentrated data clusters.

Table 4-3. Number of Observations Classified by Tier and Percentage of Observations Classified as Tier 1 or Tier 2 by Time Period and Hydrologic Region.

Tier 1	Hydrologic Region												All Data
YEAR	02	03	04	05	06	07	08	11	12	13	17	18	
1980-83	50	16	34	27	9	57	10	0	37	0	34	9	305
1984-87	55	38	15	25	8	33	3	4	14	0	69	27	324
1988-91	48	32	17	27	8	50	16	4	28	0	63	14	320
1992-95	33	18	19	27	9	45	1	0	13	0	44	32	253
1996-99	17	3	13	16	1	25	10	0	7	0	6	34	139

Tier 2	Hydrologic Region												All Data
YEAR	02	03	04	05	06	07	08	11	12	13	17	18	
1980-83	177	151	30	54	26	76	35	43	92	3	26	51	811
1984-87	147	153	42	67	17	71	42	36	121	5	64	41	889
1988-91	172	120	52	72	34	93	12	26	73	6	104	24	825
1992-95	175	122	37	83	34	84	6	18	42	6	82	55	761
1996-99	101	78	17	55	9	116	6	4	22	2	32	32	488

Tier 3	Hydrologic Region												All Data
YEAR	02	03	04	05	06	07	08	11	12	13	17	18	
1980-83	152	363	23	28	20	37	42	53	146	20	23	15	958
1984-87	191	466	32	43	22	20	47	103	159	22	23	61	1,255
1988-91	202	406	29	52	18	54	31	97	154	21	50	82	1,272
1992-95	285	372	18	66	23	45	16	51	69	28	59	24	1,074
1996-99	276	346	12	53	24	85	15	20	55	19	15	9	936

% Tier 1 or Tier 2	Hydrologic Region												All Data
YEAR	02	03	04	05	06	07	08	11	12	13	17	18	
1980-83	60%	32%	74%	74%	64%	78%	52%	45%	47%	13%	72%	80%	54%
1984-87	51%	29%	64%	68%	53%	84%	49%	28%	46%	19%	85%	53%	49%
1988-91	52%	27%	70%	66%	70%	73%	47%	24%	40%	22%	77%	32%	47%
1992-95	42%	27%	76%	63%	65%	74%	30%	26%	44%	18%	68%	78%	49%
1996-99	30%	19%	71%	57%	29%	62%	52%	17%	35%	10%	72%	88%	40%

DRAFT - Do not cite, quote, or distribute **December, 2001**
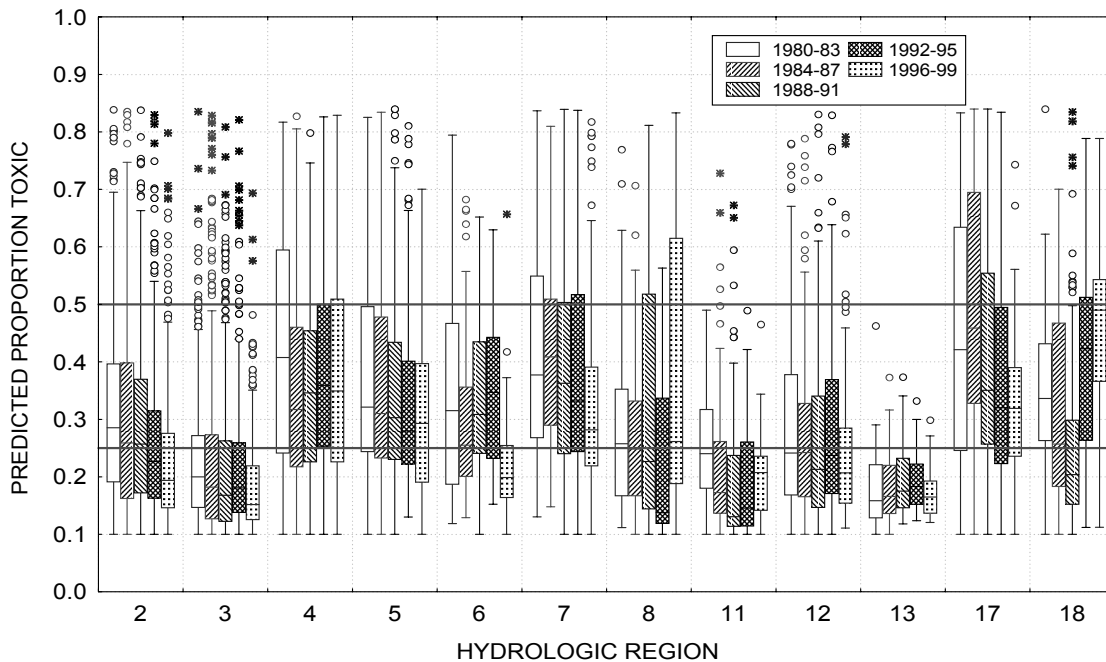
Figure 4-4. Box Plot of Predicted Proportion Toxic as a Function of Hydrologic Region for Data Clusters.
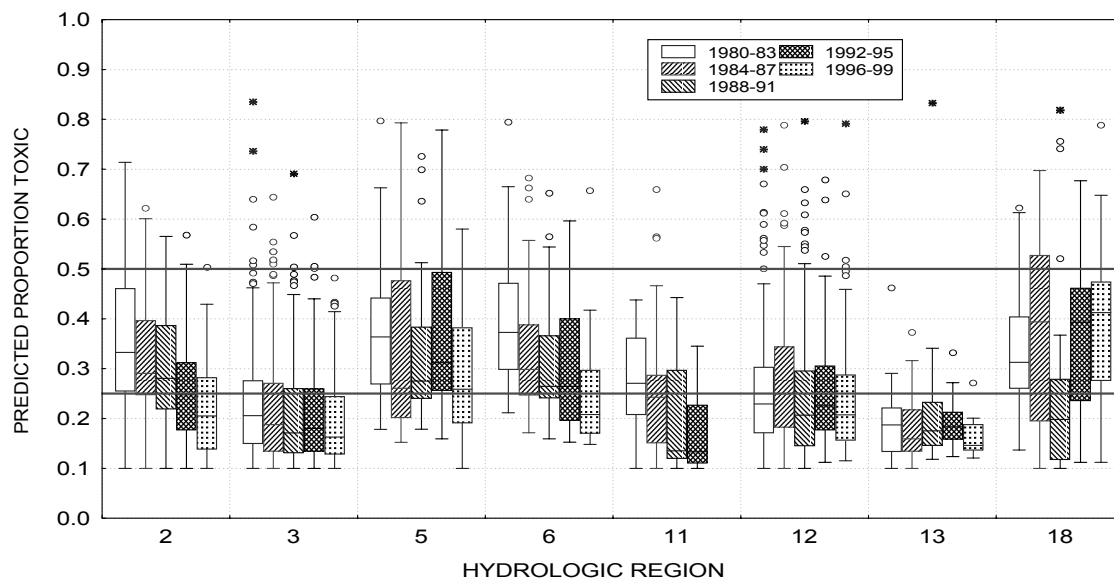

Figure 4-5. Box Plot of Predicted Proportion Toxic as a Function of Hydrologic Region for Concentrated Data Clusters.

Visual inspection of Figures 4-4 and 4-5 suggests that there may be some change in the predicted proportion toxic in some hydrologic regions and between some time periods. Two types of statistical tests are performed to evaluate these potential changes: the paired *t*-test and the Kolmogorov- Smirnov goodness of fit test. The paired *t*-test was selected to compare each combination of time periods for a shift in mean. Applying a two-sided paired *t*-test ($\alpha = 0.05$), it is possible to test for statistical

differences between the different time periods. For example, it is possible to determine whether the mean predicted proportion toxic for 1984-1987 is the same as the mean predicted proportion toxic for 1980-1983 in hydrologic region 02. For each hydrologic region, there are 10 combinations of time periods (1984-1987 vs. 1980-1983, 1988-1991 vs. 1980-1983, ..., 1996-1999 vs. 1992-1995). The Kolmogorov-Smirnov two-sample (KS) test ($\alpha = 0.05$) is used to compare an entire distribution, unlike the *t*-test, which is a comparison of means. The KS test was used to compare the first and last time periods for each hydrologic region.

In applying the paired *t*-test to all paired data, it was determined that of the 10 pairwise time period combinations, 9 showed a significant downward trend in the predicted proportion toxic for the data clusters. Eight of the 10 comparisons showed a significant downward trend in the predicted proportion toxic for the concentrated data clusters. Applying the KS test to all the data from the first and last time periods (1980-1983 vs. 1996-1999) showed a significant difference in the predicted proportion toxic between the two time periods for both the data clusters and the concentrated data clusters. The results of both of these tests suggest that sediment contamination (as measured by the predicted proportion toxic using the logistic model) is decreasing with time for all of the hydrologic regions evaluated. When looking at each hydrologic region, with sufficient data for the concentrated data clusters, the results suggest that there has been a slight decrease (Hydrologic Regions 02, 03, 05, 06, and 11) to no change (Hydrologic Regions 12, 13, and 8) in the levels of sediment contamination (as measured by the predicted proportion toxic using the logistic model). The results for individual hydrologic regions are presented in Table 4-4.

Table 4-4. Summary of Statistical Tests Used to Compare Predicted Proportion Toxic Within Hydrologic Regions.

Hydrologic Region	Data Cluster				Concentrated Data Cluster			
	Paired *t*-test results for pairwise comparison of time periods			KS Test	Paired *t*-test results for pairwise comparison of time periods			KS Test
	Increasing	Decreasing	No change		Increasing	Decreasing	No change	
02	0	8	2	decrease	0	8	2	decrease
03	0	4	6	decrease	0	6	4	decrease
04	1	4	5	no trend				
05	0	2	8	decrease[a]	0	7	3	decrease
06	0	5	5	decrease[a]	0	7	3	decrease
07	0	6	4	decrease				
08	1	2	7	no trend				
11	0	6	4	decrease[a]	0	3	3	decrease[b]
12	0	7	3	decrease[a]	0	5	5	no trend
13	0	1	9	no trend	0	1	9	no trend
17	0	1	9	decrease[a]				
18	5	1	4	increase[a]	5	1	4	no trend
Overall	0	9	1	decrease	0	8	2	decrease

[a] There is a significant difference between the distributions from the two time periods; however, the cumulative density function overlaps. The result presented in the table corresponds to whether the maximum difference indicates a decrease or an increase in concentration. Conservatively, these results should be considered "inconclusive."

[b] Comparison performed between 1980-1983 and 1991-1995 data because of lack of 1996-1999 data.
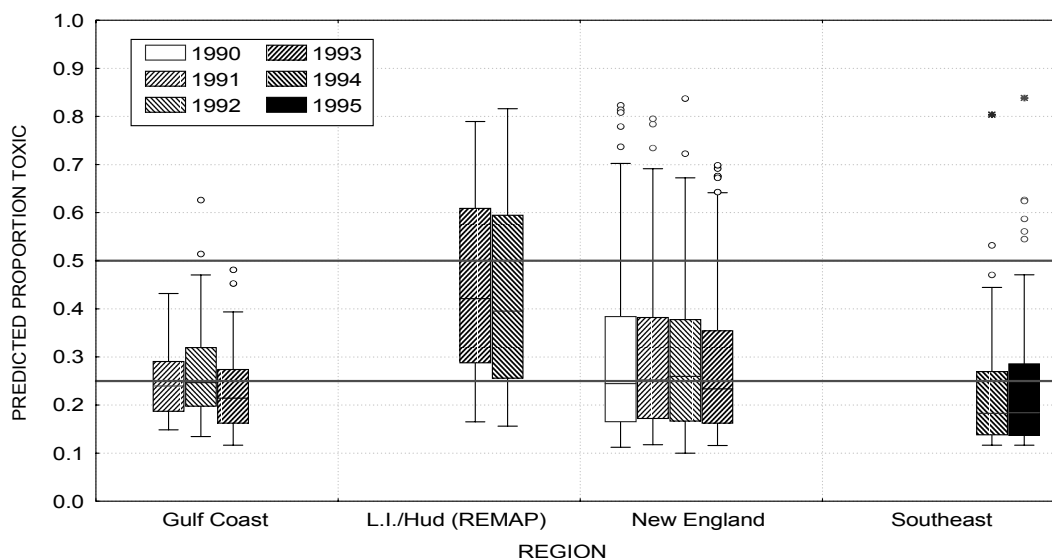
Figure 4-6. Box Plot of Predicted Proportion Toxic as a Function of Region for EMAP Data.

Box plots of the predicted proportion toxic associated with EMAP data are presented in Figure 4-6 for the four major regions where EMAP data were available. Using an ANOVA test, it was determined that there were no significant differences in predicted proportion toxic for different years of data compiled in Long Island/Hudson (L.I./Hud), New England, and Southeast regions. There is a significant difference (at the 95 percent confidence level) in predicted proportion toxic for different years of data collected along the Gulf Coast. Using a two-sample *t*-test, it was determined that the mean predicted proportion toxic in 1993 is less than the mean predicted proportion toxic in either 1991 or 1992, but the mean predicted proportions toxic in 1991 and 1992 are not significantly different.

Sediment Core Analysis

The United States Geological Survey (USGS) National Water-Quality Assessment (NAWQA) program is reconstructing water-quality histories using lake and reservoir sediment cores. The approach used for this assessment is paleolimnology - the use of age-dated sediment cores to reconstruct water quality histories using radioactive tracers and physical markers in the cores such as the pre-reservoir soil boundary in the reservoirs. Using this approach, trends in concentrations of numerous hydrophobic organic compounds (Eisenreich et al., 1989; Hites et al., 1981; Van Metre et al., 1997; 2000) and trace elements (Callender and Van Metre, 1997) have been identified in a variety of settings.

The NAWQA program is using paleolimnology methods to determine trends in metals and hydrophobic organic compounds in river basins (Callender and Van Metre, 1997; Van Metre et al., 1997). Sediment cores were collected from 1996 to 2000 in 15 reservoirs and 7 natural lakes located in or near 15 U.S cities (Table 4-5). Sites were chosen based on watershed land use to represent one of three general land use settings: older urban development (developed prior to about 1940s) dominated by residential and commercial land uses; newer urban development characterized by rapid urbanization beginning in the 1950s or later; and reference sites with little or no development. Land use in the watersheds of these reservoirs and lakes ranges from undeveloped and protected forest to nearly 100 percent urban.

Table 4-5 Sediment core locations

Name	Location (major urban areas)
Lake Anne	Washington, DC
Lake Ballinger	Seattle, WA
Berkeley	Atlanta, GA
R.R. Canyon Lake	Los Angeles, CA
Great Salt Lake, Farmington Bay	Salt Lake City, UT
Lake Harriet	Minneapolis, MN
Lake Hemet	Los Angeles, CA
Lake Houston, South	Houston, TX
Lake Killarney	Orlando, FL
Lowrence Creek Lake	San Antonio, TX
Lake Mead, Las Vegas Bay	Las Vegas, NV
Newbridge Pond	New York City, NY
Orange Reservoir	Newark, NJ
Packanack Lake	Newark, NJ
Panola	Atlanta, GA
Palmer Lake, West Lobe	Minneapolis, MN
Sloans Lake	Denver, CO
Sand Lake	Orlando, FL
Town Lake	Austin, TX
Lake Washington	Seattle, WA
White Rock Lake	Dallas, TX
West Street Basin	Los Angeles, CA

Contaminants analyzed in this program included major and trace elements (including arsenic, lead, mercury, and zinc), organochlorine pesticides (including DDT, chlordane, and dieldrin), polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs). Van Metre and Mahler (2001, in preparation) describe specific methods used for sediment collection, age dating, and the chemical analysis.

In this analysis, trends in concentrations of individual constituents were evaluated using the Spearmans rank correlation test to determine if there was a statistically significant relationship between concentration and time from 1975 to the top of the core. Trends were also evaluated by computing the percent change in the mean concentration from the 1970s (1970-1980) to the 1990s (1990 to the top of the core). The starting times for these evaluations, 1975 and the decade of the 1970s, were chosen for a variety of reasons including the passage of significant environmental legislation and establishment of new programs (e.g., the establishment of the U.S. Environmental Protection Agency in 1971). Other reasons for selecting this timeframe include the promulgation and enforcement of regulations under the Clean Water and Clean Air Acts and other actions such as: the banning of DDT in 1972; restrictions of PCBs in 1971 and their banning in 1979, and; the introduction of unleaded gasoline in the early 1970s. This time frame also corresponds to the availability of consistent, digital, national land use data which

are available only for the 1970s (GIRAS) and 1990s (MLRC). Trends in total DDT were evaluated from 1965 to the tops of the cores because DDT use in the U.S. peaked in the early to mid 1960s.

Trends in concentrations of eight trace elements, nine individual PAHs and total PAH (the sum of 18 two- to seven-ringed parent PAH and their alkylated homologues, excluding perylene (Van Metre et al, 2000), 5 organochlorine pesticides, and total PCBs were tested. These elements and compounds were chosen for analysis because they all have sediment quality guidelines (McDonald et al., 2000) and include all of the constituents measured by NAWQA studies that have pronounced temporal trends in multiple sites (Callender and Van Metre, 1997; Van Metre and Callender, 1997; Van Metre et al., 1997; Van Metre et al., 2000).

The rank correlation test has at least two limitations as applied here. First, the data sets are small, averaging 8 and 14 samples for organic compounds and trace elements, respectively. Second, the test is only for monotonic trend. It will not identify constituents or sites showing non-monotonic temporal variations, for example peaks or valleys within the time period tested. It should also be noted that the evaluation of a trend, as measured by the change in the mean concentration, does not provide a measure of statistical significance of change. It only is a measure of the relative magnitude of concentrations for the two time periods (Van Metre and Mahler, 2001 (in preparation)). In most cases these two evaluations of trend yield similar results. However, in a few cases opposite conclusions regarding trends are indicated by these evaluations, for example increasing mean concentrations for three PAH at Sand Lake that trend tested as decreasing. A few such differences are to be expected because the change in the mean is a parametric test (affected by extreme values) whereas the rank correlation test is non-parametric (not affected by extreme values) and the time periods differ (all of the 1970s versus a starting time of 1975).

Results

Statistically significant increasing trends in total PAH concentrations occur at 9 lakes (at 90 percent confidence; 8 at 95 percent) and significant decreasing trends at 2 (Figure 4-7). All nine lakes with increasing trends in PAH are in urban watersheds. Of the 11 lakes that did not have significant trends, 3 are reference lakes (Panola, Sand, and Hemet) and 7 of the other 8 are either in stable urban watersheds (Lakes Harriet, Killarney, Orange, and Packanak), are just beginning to urbanize (Lakes Houston and RR Canyon Lake), and/or are in high erosion settings where urban effects are diluted (Lake Mead in addition to Lakes Houston and RR Canyon Lake).

The two lakes that had significant decreasing trends are Lake Washington in Seattle and West Street Basin, a small pond in Anahiem, California. Both have older urban development in their watersheds which may be related to the relatively higher concentrations of PAH in older sediments. PAHs in some older urban areas have been shown to have decreased since highs in the 1940s-1960s (Van Metre et al., 2000). Additionally, sewage inputs to Lake Washington ended in 1967 and the Asarco copper smelter in nearby Tacoma was closed in the 1985. These changes could contribute to the decreasing trends in PAH and the closing of the smelter very likely contributes to the decreasing trends in most trace elements.
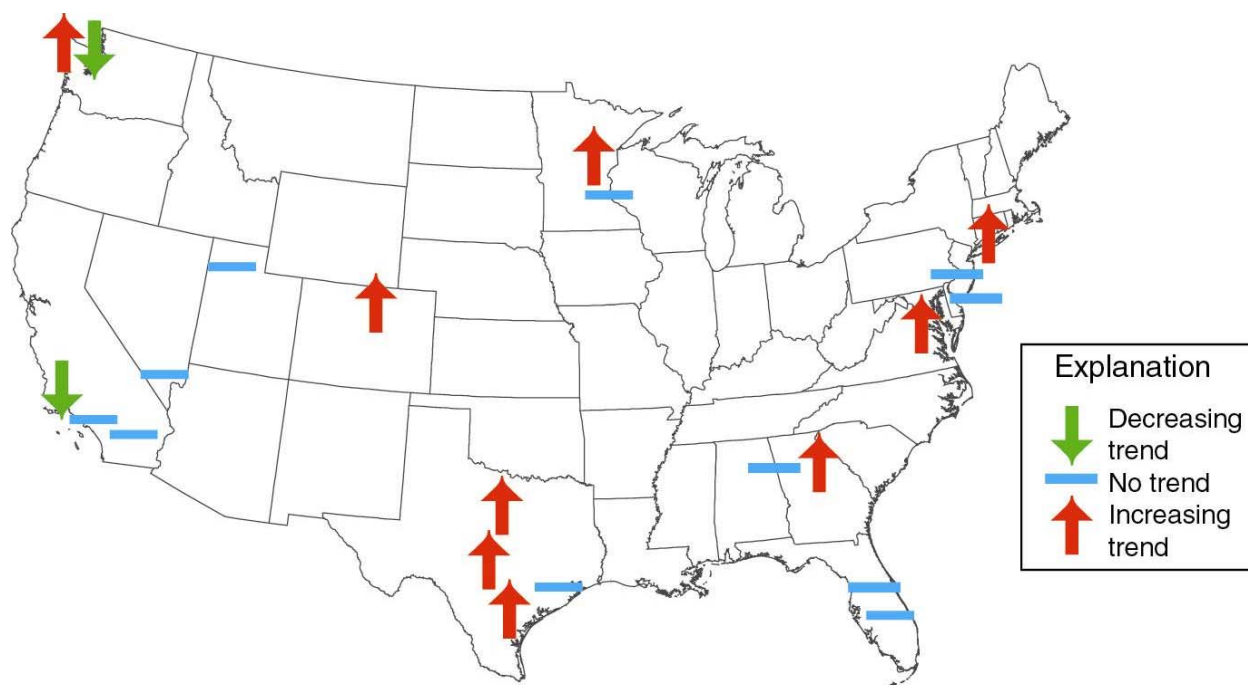
Figure 4-7. PAH trends throughout the United States

In Figure 4-8 a dramatic increase in PAH concentrations in White Rock Lake is observed since urbanization of the watershed began in the 1950s. This pattern is repeated in lakes with urbanizing watersheds across the United States (Van Metre et al., 2000). PAHs are produced by combustion of fossil fuels (oil, coal, gasoline, diesel, and wood). They have many urban sources including: industrial and power plant emissions, car and truck exhaust, tires, asphalt roads, and roofs. These sources are reflected in the relation between historical traffic data for greater Dallas and PAHs in the sediment core from White Rock Lake (Figure 4-8).

The analysis of the organochlorine compounds (pesticides and PCBs) showed that only a few had significant trends since 1975. Considering the regulatory history of these compounds, this is a little surprising. One factor that

Figure 4-8. White Rock Lake PAH concentrations

could contribute to the lack of statistically significant trends is the combination of large variability among samples and small sample size. Another that is particularly relevant for PCBs and DDT and it's metabolites is that concentrations have been decreasing exponentially in the environment since use peaked and began to decline in the 1960s (Van Metre et al, 1998). The period chosen here beginning in 1975 is after peak concentrations and on the flatter portion of the exponential decrease and may, therefore, be less sensitive to detecting trends. In fact, using the time period of 1965 to the top of the core, significant decreasing trends in total DDT occur at 12 of the 22 lakes (Figure 4-9).
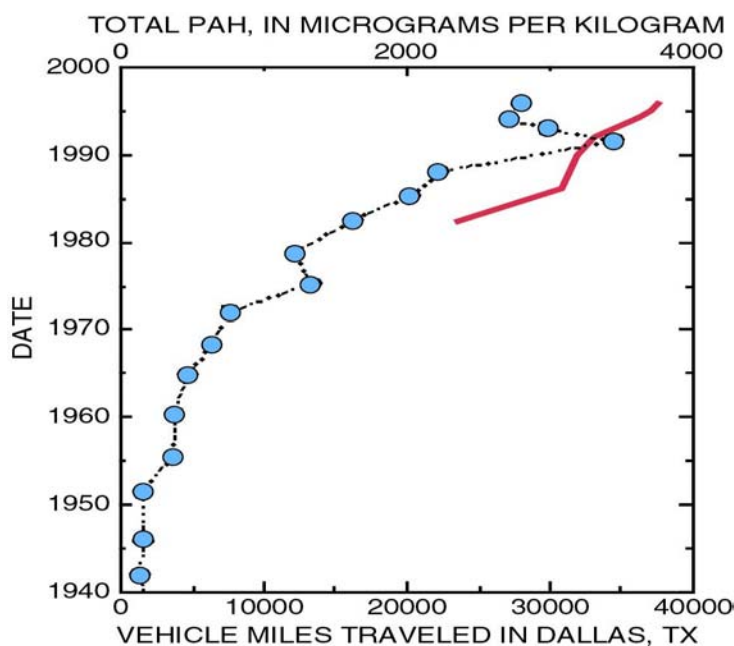
Figure 4-9. DDT trends throughout the United States

Among the organochlorine compounds it is also notable that dieldrin and chlordane have increased in almost as many lakes as they have decreased since 1975. The mean concentrations from the 1970s to 1990s for these two pesticides increased 13 times and decreased 14 times and the rank correlation test showed two decreasing trends and 3 increasing trends. These trends, and lack of trends, suggest that more recent urban use of chlordane and dieldrin have caused increasing trends in some lakes or delayed the onset of significant decreases noted elsewhere for DDT and PCBs (Van Metre et al., 1998).

DDT use was widespread in the United States in the 1950s and 1960s, and sediment cores from many lakes show a large DDT peak in the early 1960s (Van Metre et al., 1998). Trends in total DDT (the sum of DDT and its breakdown products, DDD and DDE) closely follow historical use of DDT as can be seen from the sediment core from White Rock Lake in Texas (Figure 4-10). The initial occurrence of the DDT in cores is usually in the 1940s, when widespread use began. Peak concentrations are from the late 1950s to the mid 1960s, when use peaked. DDT use declined in the 1960s and was eventually banned in 1972. Levels of DDT have been decreasing by about half every 10 years since the 1960s, although they are still at detectable levels in all but the most pristine lakes.
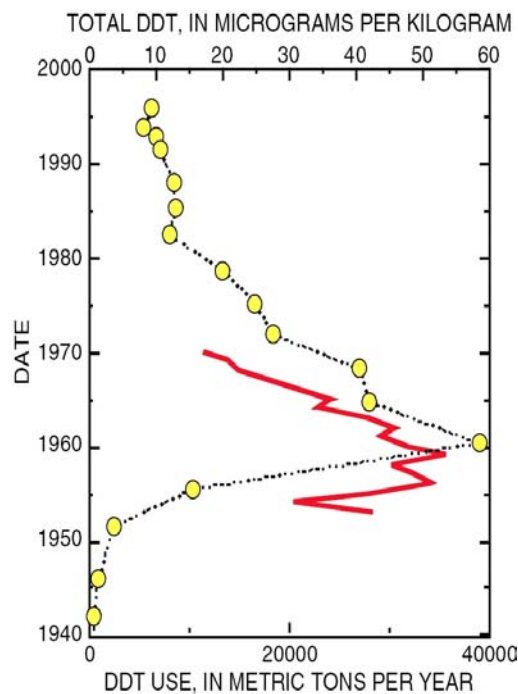

Figure 4-10. White Rock Lake DDT concentrations

The most consistent trend for any of the constituents tested is the decreasing trend in lead since the mid-1970s. All 22 lakes including the three reference lakes had statistically significant decreasing trends (Figure 4-11). Decreasing trends in lead have been reported previously and have been attributed to the large reduction in anthropogenic lead releases since the 1970s brought about by the switch to

Figure 4-11. Lead trends throughout the United States

unleaded gasoline and to a lesser extent, reductions in industrial and waste emissions (e.g. Callender and Van Metre, 1997). Significant trends in all 22 lakes, including the three reference lakes, indicates the strength of the gasoline lead signal in the U.S. environment. As can be seen from Figure 4-12, lead concentrations in the White Rock Lake sediment core track well with national atmospheric trends in lead. Two other trace elements had somewhat consistent trends; chromium and nickel each increased in only one lake and decreased in 9 and 8 lakes, respectively. The only apparent pattern to these decreases is that they mostly occurred in the central and western U.S., with the one exception of a decreasing trend in chromium in Lake Killarney in Orlando. Three other elements, arsenic, copper, and mercury, had significant trends in 10 or more lakes, all with more decreasing trends than increasing. No clear regional or land use patterns are apparent for trends in these elements, or for the four sites with decreasing trends in cadmium.
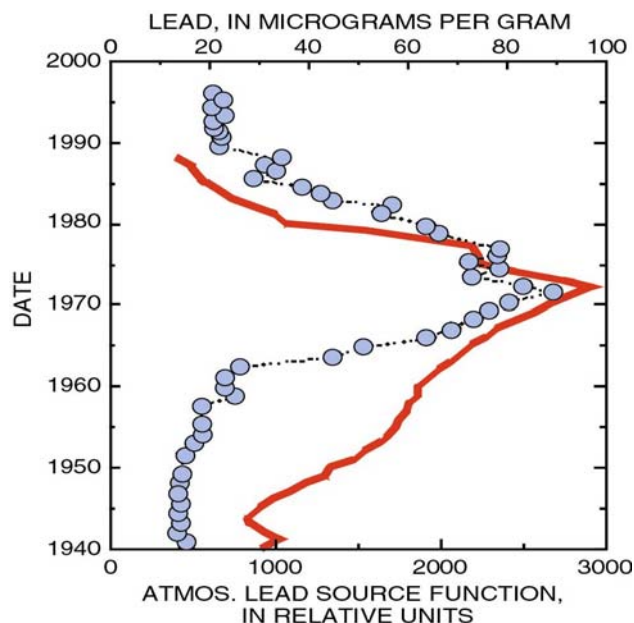

Figure 4-12. White Rock Lake Lead concentrations

The only trace element with more increasing trends than decreasing was zinc. Nine of the 19 urban lakes had increasing trends in zinc (at a 90 percent confidence level) and 4 had decreasing trends. Increasing levels of zinc in urban and urbanizing watersheds could be a result of the use of zinc in rubber tires and increasing levels of vehicle use on U.S. roads. The two lakes with decreasing trends in zinc, Lakes Washington and Harriet, are both in older urban areas. As noted above, Lake Washington had large historical inputs of metals from the Asarco smelter in Tacoma. Lake Harriet had high levels of PAHs in the 1940-1950s, possibly from home heating with coal (Van Metre et al., 2000) and has high levels of metals in the same time period in the core.

Discussion

An assessment of the contamination of sediments in lakes, rivers, and estuaries using sediment cores is a useful tool to evaluate the water quality in these waters as can be seen from the information outlined above. In addition, sediment core analysis can be used as a feedback to evaluate the impact of current legislation in reducing the level of contaminants in different waterbodies of the United States. A major limitation of the study of historical trend analysis is the post depositional disturbance of the sediment by benthic organisms and other physical processes such as wave action and dredging. Researchers have used different sediment dating techniques to study the existence and extent of such disturbances.

Some of the most significant sources of persistent and toxic chemicals have been eliminated or reduced as the result of environmental controls put into place during the past two decades. In addition, effluent controls on industrial and municipal point source discharges and best management practices for the control of nonpoint sources have greatly reduced contaminant loadings to many rivers and streams. The results of better controls over releases of sediment contamination are evident from the case studies presented.

Metals and persistent organic chemicals are the contaminants most often associated with sediment contamination. Despite recent progress in controlling sediment contaminant releases to the different compartments of the environment, active sources of these contaminants still exist. These include nonpoint source loadings such as surface water runoff and atmospheric deposition, point source loadings, and resuspension of in-place sediment contaminants from historical sources. In order to achieve EPA's Contaminated Sediment Goals, it is evident from the information presented in this chapter that a combination of pollution prevention, source control, and continuous monitoring is essential.